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(54) Name of Invention: Dry-etching Method

(57) Summary

Purpose: To etch SiO₂ layers while sustaining a high electivity ratio toward Si₃N₄ layers.

Makeup: With a dry-etching device that can create high-density plasma in which the ion density is 10^{11} ions/cm³ or more, one uses a fluorocarbon (FC) gas represented by the general formula $C_x F_y$ (with y equal to or less than $x + 2$). As the dissociation proceeds to a high degree in the gases in such high-density plasmas as ECR plasma and the like, CF^+ is yielded with good efficiency from C_6F_6 gas that could only deposit carbon-system polymers with the usual RF plasmas, making it possible to do high-speed etching of insulative film 4 between silicon dioxide layers. On the other hand, since the above-noted FC gas has a large C/F ratio, it does not produce excessive F^+ , and high selectivity is obtained toward underlying Si_3N_4 film 3. Conversely, when etching a silicon dioxide layer with the Si_3N_4 layer as a mask, high mask selectivity is obtained.

Scope of Patent Application

Application Item 1: In an etching device capable of producing plasmas of a high-density of 10^{11} ions/cm³ or more, a dry-etching method characterized by causing the production of etching-gas plasmas that are mainly fluorocarbon chemicals expressed in the general formula $C_x F_y$ (with x and y being natural numbers satisfying the relation of y being equal to or less than $x + 2$) and characterized by selectively etching the layer of silicon oxide-system material formed on a layer of silicon nitride system material.

Application Item 2: In an etching device capable of producing plasmas of a high-density of 10^{11} ions/cm³ or more, a dry-etching method characterized by producing an etching-gas plasma in which a fluorocarbon system compound expressed by the general formula $C_x F_y$ (with x and y being natural numbers satisfying the relation of y being equal to or less than $x + 2$) has as its main component the fluorocarbon compound, and characterized by etching layers of silicon oxide material while using as a mask a layer of silicon nitride material patterned in a prescribed shape.

Application Item 3: The dry-etching method described in either of Application Items 1 or 2, which is characterized by the above-noted fluorocarbon system compound being a hexafluorocarbon.

Detailed Explanation of Invention

0001 Field for Commercial Utilization: This invention relates to dry-etching methods applied in such fields as the manufacture of semiconductor devices, and in particular bears on a method of etching while securing a high selectivity rate between layers of silicon nitride-system materials and silicon oxide-system materials.

0002 Usual Technology: For the component materials of interlayer insulating films in silicon devices, silicon compound layers are generally used. Among those the most widely used is silicon oxide (SiO_x , typically with $x = 2$). At this time, the dry etching of SiO_x interlayer insulating film is a technique that has matured for application mostly to 64K DRAM production and even for mass-production lines.

0003 The above-noted dry etching has from the outset used as its etching gas mainly components of such fluorocarbon compounds as CHF_3 and CF_4/H_2 , CF_4/O_2 or $\text{C}_2\text{F}_6/\text{CHF}_3$ combinations. This is due to such reasons as (a) carbon atoms contained in fluorocarbon compounds produce a strong bonding energy between atoms on the surface of the SiO_x layer and work to break or weaken the Si-O bond; (b) they can produce CF_x^+ (typically, $x = 3$) the main source for etching SiO_2 layers; and (c) by controlling the C/F ratios (the ratio of C and F atoms) in the etching-reaction system it can optimize the amount of carbon polymer deposited and attain high selectivity toward resist masks and underlying layers.

0004 What I here call the underlying layers indicates mainly the silicon substrate, poly-silicon, polycide [sic] film, etc.

0005 Silicon nitride (Si_xN_y , especially when $x=3$ and $y=4$) also is an insulating-film substance applied in silicon devices. With dry etching of Si_xN_y layers, too, basically the same gas components are applied as in the etching of SiO_x layers. However, whereas SiO_x layers are etched mainly by the mechanism of ion-assist reactions, Si_xN_y layers are etched based on the radical-reaction mechanism making F^+ the main source of etching; and the etching rate are also is faster than with silicon dioxide layers. This can be predicted to some degree by the fact that the bonding energy relationships between atoms is Si-F (553kcal/mole) > Si-O bond (465 kJ/mole) > Si-N bond (440kcal/mole).

0006 Some differences emerge from the calculating methods in the atomic bonding energy; but here I am citing data recorded in R.C. Weast's "Handbook of Chemistry and Physics," 69th ed. (1988), CRC Press, California.

0007 Now, among the manufacturing processes for silicon devices there are several requiring high-selectivity etching between SiO_x and Si_xN_y layers. For instance, the etching of an Si_xN_y layer on top of an SiO_x layer is done under the LOCOS method in patterning to define element-separation regions. Under current conditions, where pad oxide films (SiO_2) for holding Barr's peak [Assumed from Japanese phonetics, Could also be buzz peak. -- Translator] lengths to a minimum are made as thin films, the above-noted etching is a process requiring particularly high selectivity toward the underlying SiO_x layer.

0008 Also, in recent years, as devices have become tinier and more complex, there are increasingly cases in which Si_xN_y layers are formed as etching-blocker layers in a variety of places to prevent etching damage, thus giving rise the need also for highly selective etching of SiO_x layers on Si_xN_y layers. For example, to reduce substrate damage when over-etching a thin Si_xN_y layer is interposed on the substrate surface, or a gate-insulating film having a so-called ONO structure (SiO_x layer/ $\text{Si}_x\text{N}_y/\text{SiO}_x$ layer) is formed. Or, in cases when an Si_xN_y layer is laminated on the surface of a gate electrode, the etching of an SiO_x layer done on this must be reliably halted.

0009 And yet, to do highly selective etching between laminated layers of differing substances it generally is desirable to have some degree of difference in the atomic-bonding energy of both layers. But, with SiO_x and Si_xN_y layers the atomic bonding

energies of the Si-O and Si-N are comparatively close, making it intrinsically difficult to do highly selective etching between the two.

0010 Various places have for some time been pushing the development of techniques for making selective etching possible.

0011 We have several reports on techniques for etching Si_xN_y layers on SiO_x layers. For instance, this inventor reported in Patent Report Release Sho. 61-142744 a technique for using etching gas that combines CO_2 at 30~70% mole ratio with such gases as CH_2F_2 in which the C/F ratio (ratio of carbon and fluorine atoms in a molecule) is small. Gases with small C/F ratios can yield CF_x^+ (especially when $x=3$), the etching element for SiO_x layers only by re-combination of the F^+ ; but when a large quantity of CO^+ is supplied into this system and it captures and removes F^+ in the form of COF , the amount of CF_x^+ produced is reduced and the etching rate of the SiO_2 layer declines. On the other hand, Si_xN_y can be etched with F^+ as the main etching element, so that the etching rate scarcely changes even if the CF_x^+ declines due to a large amount of CO_2 introduced. In this way, selectivity can be obtained between both layers.

0012 Also, in "Proceedings of Symposium on Dry Process" 88:7, pp. 86-94 (1987) a technique is reported for supplying NF_3 and Cl_2 into a chemical dry-etching device and using FCl formed in that atmosphere by a microwave discharge to etch an Si_xN_y layer on SiO_x . Whereas the Si-O bond includes 55% ion bonding, for the Si-N bond it is 30%, making the common [shared] bonding ratio high. I.e., the nature of the chemical bond in the Si_xN_y layer is close to that of the chemical bond (common bond) in monocrystalline silicon, and such radicals as the F^+ and Cl^+ produced by dissociation from the FCl do the etching. However, since SiO_x layers are scarcely etched by these radicals, highly selective etching becomes possible.

0013 Issues the Invention Seeks to Resolve: As discussed above, a number of techniques for selective etching of Si_xN_y layers on SiO_x layers have been reported. This is natural, in a sense, when one considers the etching rate of both layers. That is because with a process that relies mainly on radical reactions, if the SiO_x layer is exposed during the etching, the etching rate naturally slows down.

0014 However, there are problems also with the usual technology. For instance, in the above-discussed process using FCl , the anisotropic processing is essentially difficult because of using the radical reactions.

0015 On the other hand, because securing selectivity is difficult as compared to the reverse case, we have few examples of any reports. That is because even though SiO_x layers are etched by mechanisms making ion-assisted reactions primary, radicals are inevitably created in that reaction system; and at the point when Si_xN_y becomes exposed these radicals raise the etching rate of the underlying layer.

0016 Quite recently, techniques are being proposed that achieve this using a high-density plasma that reduces the amount of radicals produced by using a new plasma source. For

instance, in "Proceedings of the 43rd Symposium on Semiconductors and Integrated Circuit Technology," p. 54 (1992), there is introduced an example of a process that uses C₂F₆ (hexafluoroethane) to etch an SiO_x layer formed by the TEOS-CVD method on an Si_xN_y layer formed by the LPCVD method using an induction-coupled plasma of C₂H₆ gas. In high-density plasma, gas dissociation proceeds at a high rate, so that it is thought that the C₂F₆ is nearly all broken down into small CF⁺ ions and that this contributes to the etching. Also, carbon atoms in the low C/F ratio fluorocarbon-system polymer deposited at this time couple more easily to the atoms in the SiO_x than do the nitrogen atoms in the Si_xN_y, so that they are removed at the surface of the SiO_x layer but deposit on the Si_xN_y. This is thought to be the mechanism that attains selectivity.

0017 This technique is rather promising, but falls short in not readily achieving stable selectivity. For example, the selectivity ratio in the above-discussed process is reported to be very large on the flat areas, but 20 or more in the corners. The scatter of such selectivity on the surfaces is considered to be due to the contribution of the F⁺ produced as a result of C₂F₆ dissociation proceeding quickly. So, this invention has the goal of providing a method of dry-etching SiO_x layers that can secure a stable high selectivity ratio for Si_xN_y layers.

0018 Means to Resolve the Issue: This invention's dry-etching method is proposed after reflecting on the above-described goal and is one which, in an etching device capable of creating a plasma with an ion density of 10¹¹ ions/cm³ or more, selectively etches SiO_x-system substance layers formed on Si_xN_y-system substance layers by creating etching-gas plasmas that are mainly fluorocarbon-system compounds expressed by the general formula C_xF_y (with x and y being natural numbers and satisfying the relationship of y being equal to or less than x+2).

0019 What I here call high-density plasma is a plasma made by some means or another to increase the frequency of collisions between electrons and atoms compared to the usual type of plasma. What I mean by the usual type of plasma is that, for instance, which is excited by impressing RF power between parallel plate electrodes to cause a glow discharge or by supplying microwave to a waveguide. By contrast, high-density plasma is that which promotes a high degree of gas dissociation by using electron cyclotron resonance based, for instance, on the interaction of microwave fields and magnetic fields, or by using microwave propagation modes in a magnetic field, called a whistler mode, thus achieving high ion density.

0020 As concrete examples of plasmas having ion densities of 10¹¹ ions/cm³ or more, one knows of ECR plasma, helicon-wave plasma, ICP (inductively coupled plasma, TCP (transformer coupled plasma); holo-anode [Assumed from Japanese phonetics.--Translator] type plasma, helical resonator plasma, etc.

0021 Also, as is evident from the general formula for the above-noted fluorocarbon-system compounds, they are unsaturated compounds. Carbon frames of either chain or annular form can meet this requisite. However, if the number of carbons is somewhat greater and in chain form, it will necessarily have to have structure of multiple over-

lapping bonds or multiple conjugated bonds, condensed rings, multiple rings, spiro-rings or ring collections.

0022 As examples of fluorocarbon-system compounds meeting such requisites, one can cite tetrafluoro-ethylene (C_2F_4), hexafluorobutadine (C_4F_6), tetrafluoro-cyclopropane ($c-C_3F_4$), hexafluoro-cyclo-butane ($c-C_4F_6$), hexafluorobenzene (C_6F_6), octafluoro-cyclo-butatriene ($c-C_7F_8$), octafluoro-bicyclo[2.2.1]-heptadien (C_7F_8), etc.

0023 This invention also is one that, in an etching device capable of producing plasmas of a high-density of 10^{11} ions/cm³ or more, produces an etching-gas plasma in which a fluorocarbon system compound expressed by the general formula $C_x F_y$ (with x and y being natural numbers satisfying the relation of y being equal to or less than x + 2) has as its main component the fluorocarbon compound and does etching of layers of silicon oxide material while using as a mask a layer of silicon nitride material patterned in a prescribed shape.

0024 This invention furthermore is one that uses hexafluorobenzene for its above-noted fluorocarbon-system compound.

0025 **Effects:** When one forms a high-density plasma with an ion density of 10^{11} ions/cm³ or more using an etching gas that is mainly a fluorocarbon-system compound, even under low pressure in comparison to usual RF plasmas, the fluorocarbon-system compound's dissociation proceeds; and large amounts of CF_x^+ (but mostly x=1) are produced with good efficiency. While being assisted by this richness of ions, the silicon-compound layer is being etched at a practicable rate.

0026 However, if large amounts of F^+ are produced along with large amounts of CF_x^+ , that will cause selectivity toward the Si_xN_y layer to drop. So, to keep excessive F^+ from forming in the plasma, in this invention one uses compounds with less C atoms than F atoms—i.e., fluorocarbon compounds with molecules whose C/F ratio is large as the main components of the etching gas. In the case of this invention, the number of F atoms y will at most be twice the number of C atoms x, so that if—to simplify things—one thinks of x number of CF^+ being produced from one molecule, a maximum of two F^+ will be produced.

0027 With this invention, since one uses such fluorocarbon-system compounds to etch SiO_x -system layers on Si_xN_y layers, there is no risk of the exposed surface of the Si_xN_y layer being exposed to high densities of F^+ . So, that means that high underlying-layer selectivity is achieved with respect to the underlying Si_xN_y layer. This principle holds exactly true also when, conversely, one is using an Si_xN_y layer patterned in a prescribed shape as the etching mask for its underlying SiO_x layer, achieving high mask selectivity.

0028 Now, I plan with this invention especially to use hexafluorobenzene (C_6F_6) as the above-noted fluorocarbon compound; because this is stable, easy to handle, has a large C/F ratio of 1, etc. There have earlier been tests on using C_6F_6 in etching layers of SiO_x -system material. But, as described for instance in Patent Report Hei.1-60938, since it

forms large amounts of CF_3^+ and CF_2^+ when used alone and interferes with the progress of the etching reaction, it has not been practical. In the above-noted Report, because of this interference with carbon-system polymers, one resolves the problem by using a gas in which CF_4 is admixed 1:1 with the C_6F_6 .

0029 However, because with this invention the C_6F_6 is dissociated into CF^+ in the high-density plasma, there is no fear of carbon-system polymers accumulating excessively. Moreover, even if the bonds between carbon & carbon should all be broken and six CF^+ were produced from each molecule, in theory not a single F^+ would be produced, and high selectivity would obtain with the Si_xN_y -system layer. Also, from the aspects of stability, control, etc., one can call the point that highly selective etching is realized with a single-gas system an advantage of this invention.

0030 **Application Examples:** Below, I will explain concrete examples of applying this invention.

0031 **Application Example 1:** This application example is a case of using magnetic-field microwave plasma etching with C_6F_6 gas to open a contact hole by etching an SiO_2 interlayer insulating film having an underlying Si_3N_4 film. I will explain this process while referring to Figure 1.

0032 As shown in Figure 1(a), one first forms Si_3N_4 under-film 3 10nm thick, for instance by PCVD, on silicon substrate 1 on which impurity diffusion region 2 has been formed in advance, and then uses CVD at normal pressure to form SiO_2 interlayer-insulating film 4 1000nm thick. One further applies onto above-noted SiO_2 interlayer insulating film 4 the Novolak [Assumed from Japanese phonetics.-- Translator] positive photo resist TSMR-V3 (commercial name, from Tokyo Oyo Kagaku Co.) and uses i-beam lithography and alkali development to form resist mask 5 having $0.35\mu\text{m}$ diameter opening 6.

0033 This wafer is set on the wafer-mounting electrode of a magnetic-field microwave plasma etching device and, for instance under the conditions listed below, has its SiO_2 interlayer insulating film 4 etched.

C_6F_6 flow rate:	20 SCCM
Gas pressure:	0.65 Pa
Microwave power:	1500W (2.45GHz)
RF bias power:	200W (800 kHz)
Wafer-mount temp.	20° C

0034 Here, the C_6F_6 dissociation proceeds by applying high microwave power and doing an ECR discharge, creating a high-density plasma on the order of an ion density of 10^{11} ions/cm³. In the above-noted etching process the etching of SiO_2 interlayer insulating film 4 proceeds at a high rate due to the large amount of CF^+ produced in this high-density plasma. Also, even at the point when underlying Si_3N_4 film 3 is exposed, high

selectivity toward this film is attained because excessive F^+ is not produced in the plasma. Of course, the selectivity also is good for resist mask 5.

0035 This etching resulted in contact hole 7 being formed with an anisotropic shape as shown in Figure 1(b) without corroding Si_xNy under-film 3 exposed on its under surface.

0036 In this etching process, some accumulation of carbon-system polymer, not illustrated, can be seen. This polymer is removed by the combustion effect of oxygen atoms sputtered out from the etched region of SiO_2 interlayer insulating film 4; but this contributes to protecting the surface of resist mask 5, the sidewalls of contact hole 7 and the exposed surface of Si_3N_4 under-film 3.

0037 In any case, however, it does not obstruct the etching progress by accumulating large amounts of carbon-system polymer as in the usual case.

0038 Next, the wafer is transferred to a plasma ashing device, and oxygen-plasma ashing is done under the usual conditions, removing resist mask 5 as shown in Figure 1(c). At this time the carbon-system polymer, not shown, that contributed to protecting surfaces and sidewalls is simultaneously removed.

0039 Finally, the wafer is immersed in hot phosphoric acid and under-film Si_3N_4 exposed at the bottom of contact hole 4a is dissolved and removed, as Figure (d) shows. Through the above-noted processes one can form contact hole 7, with a good anisotropic shape without damaging impurity diffusion region 2 or causing particle contamination.

0040 Application Example 2: This application example is one done by using an ICP etching device and C_6F_6 gas to etch SiO_2 interlayer insulating film in the self-aligning contact process for forming the connections of a memory node and the gate electrode of a TFT for the negative charge of an SRAM. I will explain this process while referring to Figure 2.

0041 Figure 2(a) shows the makeup of the wafer used as an etching sample in this application example. This wafer is one on which gate oxide film 13 is formed on silicon substrate 11 by surface oxidation, and on this are patterned two gate electrodes 16 of the driver transistor and Si_3N_4 etching-stopper layer 17 for protecting these gate electrodes 15 from a later etching process. The above-noted gate electrodes 16 consist—in order from their lowest layer—of polysilicon layer 14 and tungsten silicide (Wsi_x) layer 15 laminated with a tungsten polycide film. On both sides of gate electrodes 16, sidewalls 18 of SiO_2 are formed by an etching process; and impurity diffusion region 12 is formed with an LDD structure in silicon substrate 11 by two ion injections using above-described gate electrodes 16 and sidewalls 18 as a mask.

0042 SiO_2 interlayer insulating film 19 is deposited, for instance by the CVD method, over the entire surface of this wafer, and resist mask 20 is formed in a prescribed pattern on top of that. This resist mask 20 covers part of both gate electrodes 16 and has an

opening 21 at their middle region. Inside this opening 21, SiO₂ interlayer insulating film 19 is etched as far as the impurity diffusion region to form a contact hole.

0043 The above-noted wafer is set in an ICP etching device and, as one case, SiO₂ interlayer insulating film 19 is etched under the following conditions:

C ₆ F ₆ flow rate:	20 SCCM
Gas pressure:	0.65 Pa
RF source power:	2500W (2 kHz)
RF bias power:	50W (1.5 MHz)
Wafer-mount temp.	0° C

In this process the etching proceeds on SiO₂ interlayer insulating film 19 with a CF⁺ etching agent from high-density plasma on the order of 10¹¹ ions/cm³ created in an ICP etching device. As shown in Figure 2(b), the result is that SiO₂ interlayer insulating film 19 and part of gate SiO₂ film 13 are removed and contact hole 22 is completed, while still another sidewall 19a is formed on the previously formed sidewall 18.

0044 Now, during this etching Si₃N₄ etching-stopper layer 17 becomes exposed; but—as already explained for Application Example 1—the dissociation of the C₆F₆ proceeds at a high rate and the amount of F⁺ produced in the plasma is small, so that an adequately large selectivity is sustained toward Si₃N₄ etching-blocker layer 17. Hence, one is able to avoid damage to gate electrodes 16 even when etching the high steps of SiO₂ interlayer insulating film 19.

0045 Comparative Example: Here, I will explain, as an example for comparison with Application Example 2, a case of using the ICP etching device and C₆F₆ gas to form self-aligning contacts of the same SRAM. First, one sets the same kind of wafer as that shown in Figure 2(a) into the ICP etching device and etches SiO₂ interlayer insulating film 19 under the following conditions, as one example:

0046

C ₆ F ₆ flow rate:	20 SCCM
Gas pressure:	0.65 Pa
RF source power:	2500W (2 kHz)
RF bias power:	50W (1.8 MHz)
Wafer-mount temp.	0° C

0047 In this process, CF⁺ of course is created through high-efficiency dissociation, while F⁺ is produced at the same time, reducing the selectivity toward Si₃N₄ etching-stopper layer 17. As shown in Figure 3, the result is that gate electrodes 16 under the corroded Si₃N₄ etching-stopper layer 17b also are partially corroded, and the cross-sectional shapes of sidewalls 18b and 19b each deteriorated.

0048 In the foregoing I have explained this invention on the basis of two application examples; but this invention is not limited to these examples. For example, with the

above-discussed application examples I took up the ICP device and ECR plasma as the high-density plasma; but one might also use any plasma such as a holo-anode type plasma with 10^{12} ions/cm³, a helicon-wave plasma or TCP that have been reported with an ion density on the order of $10^{12}\sim 10^{13}$ ions/cm³.

0049 Using as the etching gas the fluorocarbon-system compound C_xF_y also is not limited to the above-discussed C_6F_6 , but could consist of any compound that meets the condition of y being equal to or less than $x + 2$, also being capable of existing stably and also being easily introduced into the etching chamber in a gaseous state.

0050 Again, as other processing examples that may be applied there are such ones as the etch-back of an SiO_2 interlayer insulating film on a triple-layer gate insulating film having an ONO structure ($(SiO_x/Si_xN_y/SiO_x)$). This is a process for forming sidewalls on the side surfaces of gate electrodes, and in such case the etch-back can be halted with high selectivity on Si_3N_4 film between the gate insulating films.

0051 Needless to say, the etching conditions and etching devices can be appropriately altered.

0052 Effectiveness of Invention: As is clear from the above explanations, high C/F-ratio fluorocarbon compounds that in the past could not be used because they produced too much carbon-system polymer can, with the dry-etching method of this invention, be dissociated to a high degree in high-density plasmas by using them as the main component of the etching gas. Moreover, such fluorocarbon-system compounds do not excessively produce F^+ under the conditions for discharge dissociation so that it becomes possible to achieve selective etching between the SiO_2 material layers and Si_xN_y material layers.

0053 Consequently, this invention is applicable to the manufacture of semiconductor devices that are highly integrated and high function, designed on the basis of minute design rules, and so has a very great value in their industry.

Simple Explanation of Figures

Figure 1 is a schematic cross-sectional diagram showing in processing order an example of applying this invention to contact-hole processing. Shown respectively are (a) the situation where a resist mask has been formed on an SiO_2 interlayer insulating film, (b) the situation where the etching of the SiO_2 interlayer has stopped at the underlying Si_xN_y , (c) the situation where the resist mask has been removed by ashing, and (d) the situation where the underlying Si_3N_4 film has been removed by ashing.

Figure 2 is a schematic cross-sectional diagram showing in processing order an example of the process of applying this invention to processing a self-aligning contact. Shown respectively are (a) the situation where the SiO_2 interlayer insulating film has been formed covering two gate electrodes having an Si_xN_y etching-stopper layer, and (b) the situation where a contact hole has been formed while forming sidewalls.

Figure 3 is a schematic cross-sectional diagram showing, in a comparative example of an SRAM's self-aligning contact, the situation where selectivity toward the Si_xN_y etching-stopper layer has declined and the cross-sectional shape of the gate electrodes and sidewalls has deteriorated.

Explanation of Keying Symbols

- 1, 11 ... Silicon substrate
- 2, 12 ... Impurity diffusion region
- 3 ... Underlying Si_3N_4 film
- 4, 19 ... SiO_2 interlayer insulating film
- 5, 20 ... Resist mask
- 6, 21 ... Opening
- 7, 22 ... Contact hole
- 13 ... Gate oxide film
- 16 ... Gate electrode
- 17 ... Si_3N_4 etching-blocker layer
- 18, 19 ... Sidewalls